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Thermo-mechanical and neutron lifetime modeling and design of Be pebbles in the neutron multiplier for the LIFE engine

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Nuclear Fusion

Thermo-mechanical and neutron lifetime modeling and design of Be pebbles in the neutron multiplier for the LIFE engine

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Abstract: Concept designs for the laser-initiated fusion/fission engine (LIFE) include a neutron multiplication blanket containing Be pebbles flowing in a molten salt coolant. These pebbles must be designed to withstand the extreme irradiation and temperature conditions in the blanket to enable a safe and cost-effective operation of LIFE. In this work, we develop design criteria for spherical Be pebbles on the basis of their thermo-mechanical behavior under continued neutron exposure. We consider the effects of high fluence/fast flux on the elastic, thermal and mechanical properties of nuclear-grade Be. Our results suggest a maximum pebble diameter of 30 mm to avoid tensile failure, coated with an anti-corrosive, high-strength metallic shell to avoid failure by pebble contact. Moreover, we find that the operation temperature must always be kept above 450°C to enable creep to relax the stresses induced by swelling, which we estimate to be at least 16 months if uncoated and up to six years when coated. We identify the sources of uncertainty on the properties used and discuss the advantages of new intermetallic beryllides and their use in LIFE's neutron multiplier. To establish Be-pebble lifetimes with improved confidence, reliable experiments to measure irradiation creep must be performed.

I. Introduction and background.

Laser inertial fusion-fission energy (LIFE) engines are being considered at Lawrence Livermore National Laboratory (LLNL) to produce nuclear energy from natural or depleted uranium without isotopic enrichment, and from spent nuclear fuel from light water reactors (LWR) without chemical separation into weapons-attractive actinide streams. The LIFE concept relies on a point source of 14-MeV neutrons produced by an inertial-confinement fusion (ICF) engine enveloped by a Be neutron multiplier and a sub-critical fuel blanket. The objective is to achieve ultra-deep (>99%) burn-up of the fertile or fissile fuel in the fuel blanket without extraction or reprocessing to minimize nuclear waste and mitigate nuclear proliferation concerns [Moses2008]. Among the solid fuel designs, one involves burning fertile material such as spent nuclear fuel (SNF) or depleted uranium (DU) and building up the actinide inventory which is then consumed to very high burn-ups. LIFE also provides an attractive pathway to burn fissile materials such as excess weapons-grade Pu (WG-Pu), which can achieve over 99% FIMA (fraction of initial metal atoms) in much shorter time scales [Moses2008]. The proposed LIFE engine configuration is shown in **Figure 1**. More details about the nuclear aspects, and the thermal-hydraulic and mechanical design of the reactor can be found in reference [Abbott2008].

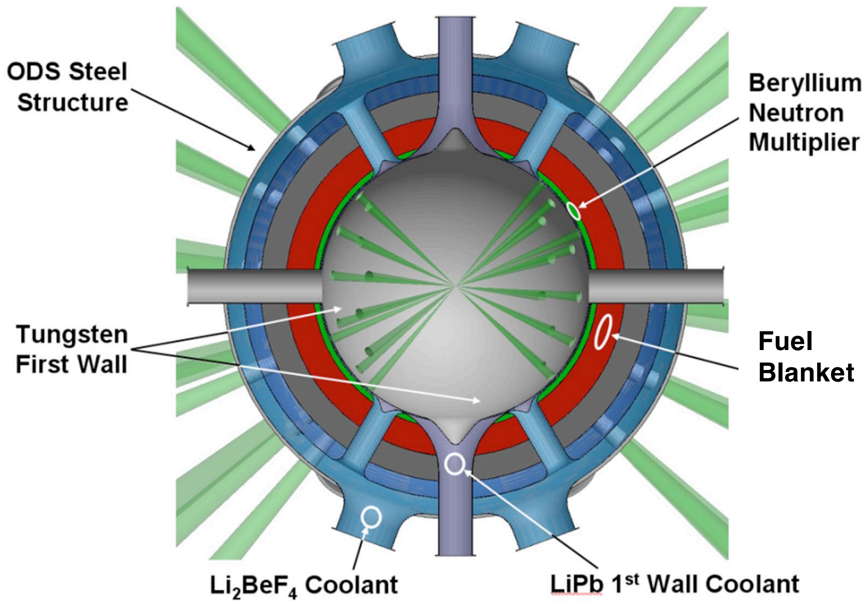

Figure 1: LIFE engine configuration showing the location of the Be multiplier.

Before striking the fuel region, the fusion neutrons first traverse a layer of Be, where they undergo moderation and multiplication via (n,2n) reactions. The Be blanket design proposed is similar to those implemented in experimental fusion breeder reactors [Moir1983], in which a large number of small (≈1 cm) pebbles are suspended in liquid Li_2BeF_4 ('flibe').

However, the neutron flux and fluence, and the coolant temperatures under which the Be pebbles have been designed to perform in the LIFE are significantly more stringent than in existing or past fusion breeders. The estimated temperature range in the neutron multiplier is 600 to 750°C, with fast fluences of up to 9.1×10^{23} n·cm^{-2} [Kramer2008]. Thus, Be-pebble lifetime estimates obtained from previous experimental campaigns at temperatures of typically 450°C and fast fluences of $\approx1.2\times10^{23}$ n·cm^{-2} corresponding to fluxes ≪14 MeV [Miller1983] are not applicable for LIFE. **Figures 2a** and **2b** show, respectively, the neutron flux at the beginning and end of life (BOL, EOL), and the fluence as a function of lifetime, averaged over the Be blanket in the DU and WG-Pu versions of the LIFE reactor (for specific fuel compositions we refer the reader to [Moses2008, Kramer2008].

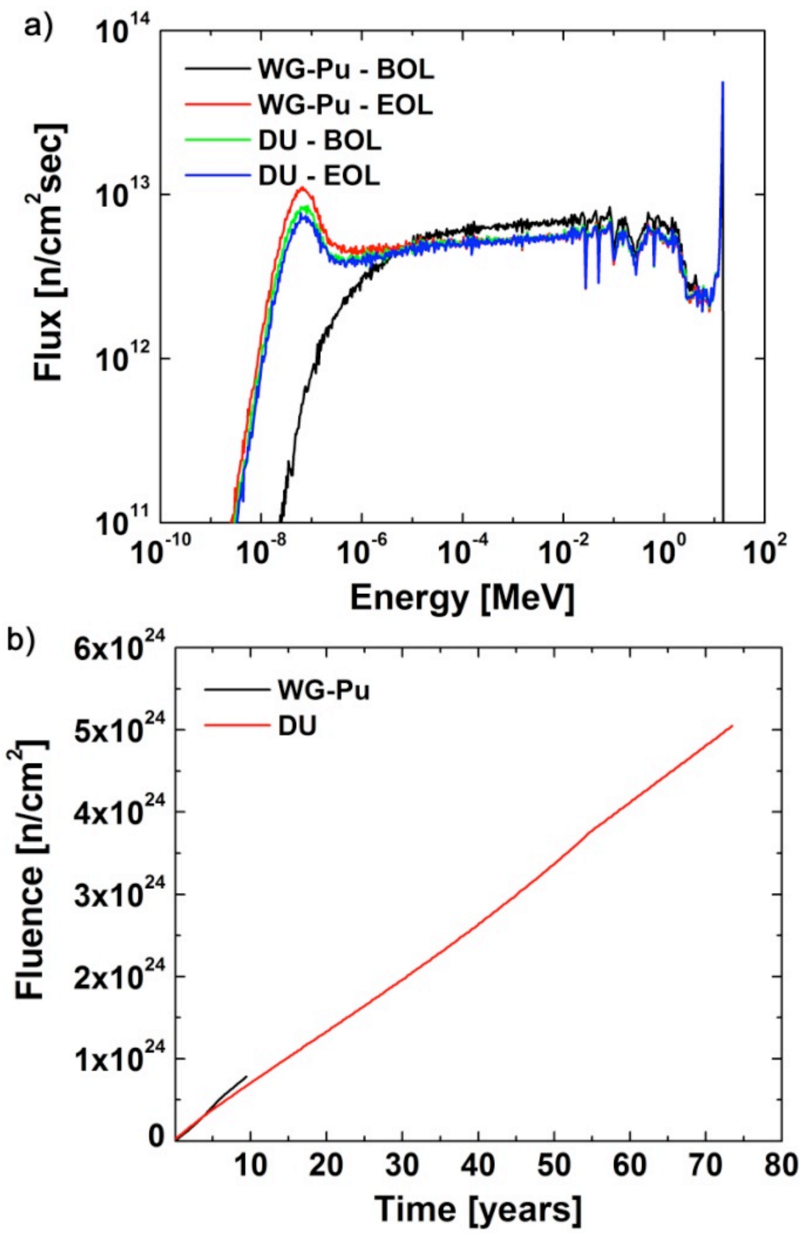

Figure 2: (a) Total fluxes at BOL and EOL in the Be multiplier for the DU and WG-Pu LIFE concepts considered here. (b) The corresponding accumulated total fluences.

Table I gives the expected operation lifetimes of both engines, and the associated fluences and damage accumulation in the Be multiplier[1].

[1] Calculated with the SPECTER code [Greenwood] using a threshold displacement energy of 31 eV.

Table I: Irradiation parameters in the two LIFE concept designs considered here.

	DU	WG-Pu
Lifetime for 99% FIMA [yr]	73.7	9.4
Total Be blanket fluence [n·cm^{-2}]	5.1×10^{24}	7.8×10^{23}
Be damage at EOL [dpa]	759.4	98.9
He accumulation at EOL [appm]	410912	52394
H accumulation at EOL [appm]	2917	370

Although the integrated damage in the DU engine is far too large to be withstood by any nuclear-grade Be, frequent component and blanket replacements during operation are proposed in LIFE. In this fashion, the aggregate damage and gas accumulation during an operation cycle becomes tolerable. Nevertheless, component replacements in LIFE may present important challenges from an engineering point of view. He accumulation is substantial in the DU engine, with >40% of the Be being transmuted into He at EOL. Tritium (H) accumulation, on the other hand, represents only ≈0.7% of the total gas storage in the Be. Experiments show that most of the tritium resides in the He bubbles, and is released above 600ºC when the pebble percolation limit is reached [Longhurst1990]. Furthermore, compared to the surrounding flibe, the tritium production rate in the Be pebbles is likely to be insignificant.

The lifetime of Be pebbles is governed by the interplay among differential thermal expansion, irradiation-induced swelling, and thermal and irradiation creep. The first two phenomena give rise to internal stresses that increase with time (fluence). Depending on the temperature, creep may relax this stress buildup before the tensile limit is reached. Several authors have suggested that, for temperatures between 400 and 500°C, thermal creep is not sufficient to relax the stresses caused by swelling, and that, approximately at 30% volumetric swelling, crumbling occurs and the particle fails [Miller1983].

In this work, we carry out a thermo-mechanical analysis of Be pebbles under LIFE design conditions using scaled swelling functions and recently obtained creep laws [Scibetta2007]. We assess the lifetime of the Be pebbles in the neutron multiplier, and recommend a particle size window based on failure and flow considerations in the Be blanket. Although large uncertainties exist for the material properties of nuclear-grade Be, and its behavior under irradiation, we have made every effort to narrow the confidence intervals at the conditions of interest. This is shown in Section III.

II. Theory

The material behavior of Be is represented by the following set of constitutive equations [Miller1993]:

$$\begin{aligned}
\varepsilon_r &= \frac{1}{E}\left(\sigma_r - 2\nu\sigma_t\right) + ct\left(\sigma_r - 2\nu_c\sigma_t\right)^n + \frac{S_r}{3} + \alpha_r T \\
\varepsilon_t &= \frac{1}{E}\left[\left(1-\nu\right)\sigma_t - \nu\sigma_r\right] + \\
&\quad + ct\left[\left(1-\nu_c\right)\sigma_t - \nu_c\sigma_r\right]^n + \frac{S_t}{3} + \alpha_t T
\end{aligned} \tag{1}$$

where ε is the strain, σ is the stress, and E, ν, and ν_c are, respectively, the modulus of elasticity, Poisson's ratio and Poisson's ratio in creep. Here, c and S are the temperature and fluence dependent creep constant and volumetric swelling, n the creep exponent, t is the time, and α is the thermal expansion coefficient. T is the absolute temperature. Eqs. (1) are formulated assuming spherical geometry and the subindices r and t (not to be confused with the time) refer to the radial and tangential (or 'hoop') components, respectively. Closure of the system of equations (1) is achieved by considering the elastic equilibrium equation $\nabla\cdot\boldsymbol{\sigma} = 0$, which for an axisymmetric solid is simply:

$$\frac{\partial\sigma_r}{\partial r} + \frac{2}{r}\left(\sigma_r - \sigma_t\right) = 0 \tag{2}$$

Solutions to these equations and their validity have been discussed by Miller *et al.* [Miller2003, Miller2008]. All the material constants in eqs. (1) are subject to change due to irradiation. Therefore time-stepping algorithms must be used to find the solution at time t_{n+1} from the state at time t_n. In the following section, we discuss the time (irradiation) dependencies of each material property separately and their relative effect on the stress evolution in the pebble. At this time, the pulsed nature of the LIFE engine is not taken into account, as preliminary studies suggest that its effect is negligible in the Be blanket [Abbott2009].

III. Material properties

Our main source of physical property data for Be is the excellent compilation by Beeston [Beeston1970], which, despite being almost four decades old, is still the reference for nuclear applications. More recently, Karditsas and Baptiste [Karditsas1995] have collected physical and structural property data as a function of temperature for magnetic confined fusion (MCF) applications, although these properties are continuously being revised [Scaffidi2000, Longhurst2003]. Gelles *et al.* have considered the effects of neutron irradiation in a comprehensive review article [Gelles1994]. Despite similar Be fabrication methods[2], these studies showcase the large variability found in the physical properties of Be provided by different manufacturers. Next, we discuss the choice of material properties used in this study under LIFE design conditions.

Thermal properties

The latest LIFE blanket design calls for a coolant temperature of 610 to 615°C in the Be multiplier. In addition, a volumetric heating of q=1.7×10^{7} W·m^{-3} due to exothermic neutron capture reactions is assumed [Abbott2008]. The temperature profile inside the particle is then given by the steady-state solution of the heat equation in an isotropic spherical particle:

$$T(r) = T_R + \frac{q(R^2 - r^2)}{6k} \qquad (3)$$

where T_R≈889K (616 °C) is the average temperature in Kelvin at the outer pebble surface [Abbott2009], R is the pebble radius, and k the thermal conductivity. The Be thermal conductivity suffers a marked decrease with increasing temperature. Beeston [Beeston1970] and Karditsas and Baptiste [Karditsas1995] give thermal conductivities of the order of 98~110 W·m^{-1}·K^{-1} at 900K, down from ≈225 W·m^{-1}·K^{-1} at 300K. With regard to irradiation effects on k, Chakin *et al.* [Chakin2001] report very radiation-insensitive thermal conductivities at temperatures >300°C up to fast fluences of the order of ~10^{23} n·cm^{-2}. Barabash *et al.* [Barabash2003] note that k only changes appreciably in conditions of significant swelling (>40%), or when a large number density of irradiation defects are present (high dose, low temperatures). They give a fluence-independent value of 125 W·m^{-1}·K^{-1} above 700°C [Barabash2003]. For consistency with all these studies, here we use a value of k=100 W·m^{-1}·K^{-1} and assume that it does not vary with fluence. For R=15-mm pebbles, for example, the temperature

[2] Almost all Be components in nuclear applications are fabricated by hot pressing of high-purity powders. The specification for nuclear-grade Be and BeO powders is provided in an updated ASTM standard [ASTM]. The rotating electrode method has also been widely used for the last decade, particularly in Europe [Cardella2004].

difference is ≈6.4°C, which results in thermal stresses of less than 12 MPa, insignificant compared to those induced by swelling, as we shall see.

Regarding the thermal expansion coefficient, both Beeston and Karditsas and Baptiste give a value of $\alpha = 1.6\times10^{-5}$ K^{-1} at 900K. To the best of our knowledge, there is no published study on irradiation effects on α in Be and here we simply use this value and maintain it constant with fluence.

Swelling

Be undergoes exothermic (n,2n) and (n,α) threshold reactions[3] (approximately 2.7 and 1.4 MeV, respectively), giving rise to large amounts of He accumulation. The formation of He bubbles, predominantly at grain boundaries and dislocation loops [Gelles1998, Kupriyanov1998, Chakin2001], results in irradiation swelling. Neutron swelling data in Be is quite limited, although recent measurements at very high fast fluences have been carried out [Chakin2001, Kupriyanov1995]. Here, we have used the phenomenological law of Beeston *et al.* [Beeston1984], obtained from seven different specimens irradiated at fast fluences between 0.7 and 1.3×10^{22} n·cm^{-2} (35 and 65 dpa), and temperatures in the 700 to 760K range:

$$S(r,F) = bF^2T^4(r) \qquad (4)$$

where b=1.83×10^{-58} is a fitting constant, F is the fast fluence (>1 MeV) in units of 10^{22} cm^{-2} and $T(r)$ is given by eq. (3). This law should be valid up to the saturation point, measured by some authors at 750°C [Leenaers2008]. The data to which eq. (4) was fitted was obtained from a series of irradiations in EBR-II [EBR-II] and it is thus not directly suitable for LIFE conditions. However, Beeston *et al.* provide a scaling procedure assuming that He accumulation is the governing swelling mechanism. Specifically, the scaling factor is computed based on a reference He production rate in EBR-II is 1540 appm per 10^{22} n·cm^{-2} fluence. Table II shows the H (≈99% tritium), He and dpa production rates for the irradiation parameters shown in Table I for both LIFE concept designs. Results have been obtained using the SPECTER code [Greenwood] and are given in Table II:

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[3] $n+{}^9Be \rightarrow {}^8Be+2n \rightarrow 2\alpha+2n$
$n+{}^9Be \rightarrow \alpha+{}^6He$
${}^6He \rightarrow {}^6Li$; ${}^6Li+n \rightarrow \alpha+{}^3H$

Table II: damage and gas production per full power year of operation in two LIFE concept designs.

	DU	WG-Pu
dpa	10.3	10.5
He [appm]	5574	5544
H [appm]	40	39
Fast fluence (>1 MeV) [$\times 10^{22}$ n·cm^{-2}]	0.87	0.90

From table II, at 10^{22} n·cm^{-2} fast fluence, the He production in the DU and WG-Pu engines is, respectively, 6407 and 6160 appm, which results in scaling factors of 6407/1540=4.2 and 6160/1540=4.0. Because this is an *equivalent fluence* factor, it is quadratic in eq. (4), *i.e.* the prefactor of the swelling law now becomes 4.2^2b=3.23×10^{-57} for the DU design, and 4.0^2b=2.93×10^{-57} for the WG-Pu case. Conversion from fast fluence to time is readily achieved through the following relations (Fig. 2b):

$$F(\mathrm{DU}) = 2.77\times10^{14}\,t$$
$$F(\mathrm{WG-Pu}) = 2.84\times10^{14}\,t$$

where t is expressed in seconds. The final swelling expressions as a function of time (s) are:

$$\begin{aligned} S_{\mathrm{DU}}(r,t) &= 2.48\times10^{-28}\,t^2T(r)^4 \\ S_{\mathrm{WG-Pu}}(r,t) &= 2.36\times10^{-28}\,t^2T(r)^4 \end{aligned} \qquad (5)$$

We assume that swelling is isotropic and, in all cases considered here, $S_r(r,t)$=$S_t(r,t)$ =$S(r,t)$. We emphasize that these expressions correspond to *volumetric* swelling, which is the reason why they are multiplied by 1/3 in eqs. (1), to convert them to linear elongation.

Several authors have suggested that swelling can be tolerated up to a maximum value of 30% (volumetric), at which either crumbling occurs [Miller 1983] or saturation is observed [Leenaers2008], although values of up to 40% have been measured at fluences of ~10^{22} n·cm^{-2} and T>1100°C [Gelles1994]. In this work, we assume that this limit results in particle failure regardless of the stress state inside the particle. Other authors have proposed that once the percolation limit is reached (≈19% porosity in the hexagonal close-packed Be lattice [Scher1970]), the He gas can be released and swelling is no longer a concern.

Creep

The last parameter to be characterized in eqs. (1) is the creep constant *c*. In this study we are interested in steady-state conditions where secondary creep is the operating creep mode. There have been several studies of thermal creep in Be pebble beds for fusion energy [Beeston1970, Billone1998, Hofer2005, Reimann2005]. However, it is well known that irradiation can both augment thermal creep and develop creep in conditions where thermal creep is absent. Moreover, the relation between creep and irradiation-induced swelling has been clearly established in a number of materials [Hassan1992]. Here, we utilize the expression for secondary creep in Be provided by Scibetta *et al.* [Scibetta2007], relating the creep rate to the existing material porosity:

$$\dot{\varepsilon}_c = A\left(\frac{\sigma_{eff}}{1-p^{2/3}}\right)^n \exp\left(-\frac{Q}{T}\right) \quad (6)$$

where A=7.21×10^{-3} s^{-1}·MPa^{-n}, Q=19470K, n=2.43, and p is the porosity. In turn, the material porosity is related to the integrated volumetric swelling by $p = \frac{S(r,t)}{1+S(r,t)}$. σ_{eff} is the effective stress, which takes into account Poisson effects in eqs. (1). It is assumed that ν and ν_c are identical and equal to 0.03.

Expression (6) links creep with irradiation and provides a good fit to previously published creep data in the temperature range of interest in this work [Kupriyanov1995, Reimann2005]. At 616°C, we have that $c = A\exp(-Q/T) = 2.22\times10^{-12}$ s^{-1}·MPa$^{-2.43}$.

Elastic and mechanical properties

The only elastic constants in eqs. (1) are the modulus of elasticity, *E*, and the Poisson ratios ν and ν_c. Here we have used a modulus of elasticity of E = 262.7 GPa at 900K [Karditsas1995]. Regarding the mechanical properties, we use an ultimate tensile strength (UTS) of 175 MPa at 616°C for S-200E Be[4] [Henshall1995], which is consistent with experimental measurements in several other Be grades [Ells1964, Moons1996]. Ells and Weir [Ells1964] have reported that the UTS is quite impervious to irradiation, especially at high temperatures.

As He and H gas builds up within the bulk, Be pebbles are expected to swell and develop an increased porosity. This effect has been studied for several Be properties, *e.g.*, it has been noted that *E* can decrease by 60% at 30% porosity [Karditsas1995]. Therefore, in this work we assume a linear relation between *E* and *p* of the form: E=E_0(1–2p), which ensures that the modulus of elasticity loses 60% of its zero-porosity value of E_0=262.7 GPa when p=0.3.

[4] Commonly used at LLNL, although Be S-65 is a higher-purity variant chosen for ITER [Moons1996].

Finally, Beeston [Beeston1970] reports that the tensile limit increases with irradiation by up to 40% at fluences of 2.4×10^{21} $n\cdot cm^{-2}$ with a tendency to saturation, although Kupriyanov *et al.* [Kupriyanov1995, 1998] have shown that this effect is almost suppressed at temperatures above 600°C.

Other important mechanical properties, not directly relevant for eqs. (1), are the yield stress and the ductility. A reasonable amount of ductility must be retained during operation for reasons that we will discuss in Section V.

IV. Results

Mechanical study of individual Be pebbles

Despite their relative simplicity, eqs. (1) contain a double (r,t) dependence in four strongly coupled terms contributing to the pebble deformation. This leads to a weak convergence formulation that requires implicit solvers and potentially small time steps.

As a 'hard' lower lifetime estimate, we plot in **Figure 3** the time at which 30% volumetric swelling is reached in both Be blankets of the DU and WG-Pu LIFE concepts at an operating temperature of 616°C. For the DU engine, a lifetime of 1.39 years –or 1.2×10^{22} $n\cdot cm^{-2}$– is obtained, which corresponds to an accumulation of 7748 He appm and 14 dpa in the pebbles. The numbers for the WG-Pu case are, respectively, 1.43 years (1.3×10^{22} $n\cdot cm^{-2}$), 7928 He appm, and approximately 15 dpa. These numbers have been calculated from eqs. (5) and Table II, and, evidently, do not reflect the true lifetime behavior of the Be pebbles, governed by eqs. (1). However, they are intended to provide a worst-case estimate assuming 30% swelling as the failure limit. As a disclaimer, we note that a failure criterion based on dpa accumulation could be equally acceptable, although there exists at this time no data above 40 dpa in Be [Kwast1996] to establish such a limit with confidence.

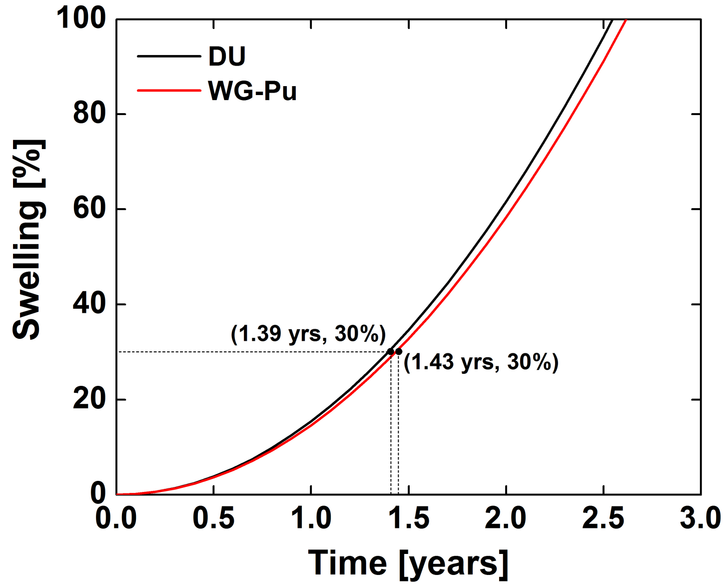

Figure 3: Swelling at 889K as a function of time in the two LIFE engine designs considered here.

Next, we compute the radial and hoop stresses in a spherical pebble by solving eqs. (1) and (2) at BOL and at 30% vol. swelling. We first disregard the effect of creep for, again, bounding purposes, *i.e.* only thermal expansion and swelling are considered. The radial dependence of the stress is given for σ_r and σ_t by solving:

$$\begin{aligned}
\sigma_r(r) &= \frac{2E}{\nu-1}\left\{\frac{1}{R^3}\int_0^R r'^2\left[S(r')+\alpha\left(T(r')-T_R\right)dr'\right]-\right.\\
&\qquad \left.-\frac{1}{r^3}\int_0^r r'^2\left[S(r')+\alpha\left(T(r')-T_R\right)dr'\right]\right\}\\
\sigma_t(r) &= \frac{E}{\nu-1}\left\{\frac{2}{R^3}\int_0^R r'^2\left[S(r')+\alpha\left(T(r')-T_R\right)\right]dr'+\right.\\
&\qquad \left.\frac{1}{r^3}\int_0^r r'^2\left[S(r')+\alpha\left(T(r')-T_R\right)\right]dr'-\left[S(r)+\alpha\left(T(r)-T_R\right)\right]\right\}
\end{aligned} \tag{7}$$

which can be solved analytically giving rise to 8th-degree polynomials in r. However, for small r, the contribution of the higher-order terms can be safely neglected, resulting in solutions of the type:

$$\sigma_r(r,R) = R^2\frac{q\left(1-\left(r/R\right)^2\right)}{15k}\cdot\frac{E\left(\alpha+\frac{4}{3}bT_R^3F^2\right)}{\nu-1}$$

$$\sigma_t(r,R) = R^2 \frac{q\left(1-2(r/R)^2\right)}{15k} \cdot \frac{E\left(\alpha + \frac{4}{3} b T_R^3 F^2\right)}{\nu - 1} \tag{8}$$

The σ_r/R^2 and σ_t/R^2 functions are plotted in **Figure 4** as a function of the ratio r/R. The curve at BOL clearly shows that thermal effects are practically negligible, consistent with the small temperature gradients that develop inside the pebble. The radial stress correctly vanishes at R, while the hoop stress changes from negative to positive at a location that varies with time. From Fig. 4, the first design criterion can be established, namely that $|\sigma| < \mathrm{UTS}$ at all times. Since the maximum hoop stress of σ/R^2=0.77 MPa·mm^{-2} is achieved for S=30% after 1.39 and 1.43 years for the DU and WG-Pu engines respectively, we have that $R^2 < \frac{\mathrm{UTS}}{0.77}$, *i.e.* R<15.1 mm. Thus, in the absence of creep, which results in stress relaxation, a pebble size R=15 mm serves as an upper bound to ensure a safe stress level within the lifetimes established by 30% swelling.

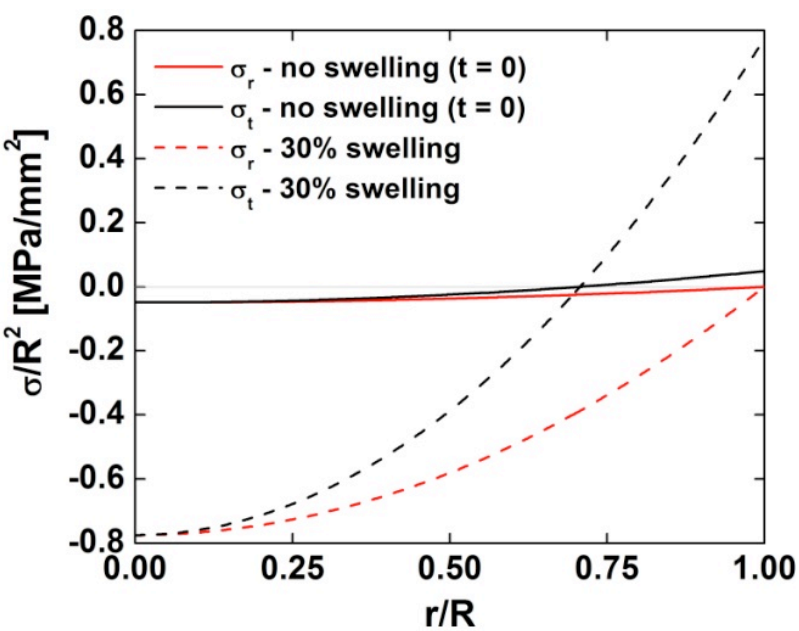

Figure 4: Radial and tangential stresses as a function of the ratio (r/R) when thermal creep is neglected.

The effect of creep is studied by solving eqs. (1) with all the terms for a R=15-mm Be pebble with, and considering all the fluence dependencies simultaneously. **Figure 5** shows the effect of creep at several relevant times for both LIFE concept designs. At zero time, the curves evidently coincide with those shown in Fig. 4, particularized for R=15 mm. As t increases, the effect observed is a decrease in the compressive stresses (both radial and tangential) at the center of the particle (r=0), and a tempering of $\sigma_t(R)$. After approximately 57 hours all the stresses practically vanish and the particle enters a fully relaxed steady state. This temporal behavior is better illustrated in **Figure 6**, where eqs. (1) are integrated in time for $\sigma_t(R)$ with and without creep. These two curves constitute lower and upper lifetime estimates for a 15-mm Be pebble in the LIFE neutron multiplier. The stress relaxation time constant of ≈57 h when creep is considered is significantly shorter than that of swelling. In other words, swelling effects manifest themselves over time scales much larger than those associated with creep. To estimate the temperature at which creep no longer dominates over swelling, we have solved eqs. (1) as a function of T and obtained a value of 450°C. Operating at temperatures below this critical value is anticipated to shrink the Be pebble lifetime significantly.

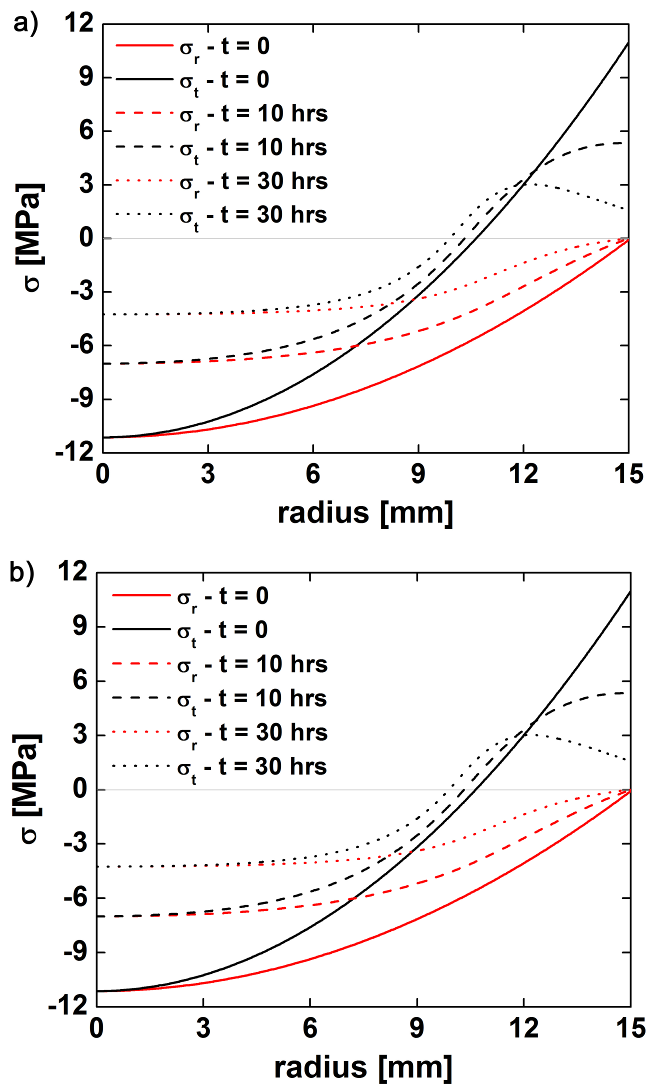

Figure 5: Radial and tangential stresses as a function of radius for a pebble with R=15 mm when thermal creep is included for (a) the DU and (b) WG-Pu engines. Stresses are shown at three different times. The gradual time softening as thermal creep sets in can be clearly observed. After approximately 57 hours, all the stresses relax practically to zero due to thermal creep.

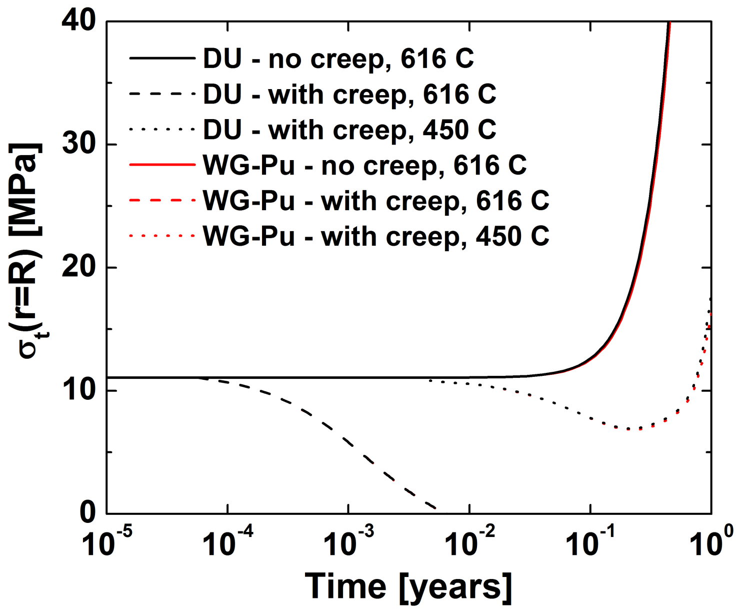

Figure 6: Thermo-mechanical evolution of an R=15-mm Be particle in the LIFE engine at two different temperatures. For T=616°C, both the upper and lower bound estimates (with creep and no creep, respectively) are shown. The curves for the DU and WG-Pu reactors with creep at 616°C are indistinguishable from one another. At T=450°C, only the case with creep has been considered since it is at this temperature when creep is no longer able to relax the stresses created by swelling.

The time evolution presented in Figure 6 corresponds to two extreme cases, namely, (i) only swelling is considered, and (ii) swelling plus thermal creep are considered. Thermal creep is extremely efficient in relaxing all the stresses in the pebble on a time scale before swelling can develop, essentially rendering the pebble lifetime infinite from a thermal-mechanical point of view. However, there are estimates that creep saturates with fluence due to a large accumulation of irradiation-produced defects. This will likely inhibit thermal creep's beneficial effects, causing the true lifetime to be somewhere in between the two curves shown in Fig. 6. From a temperature standpoint, including irradiation creep may increase the critical value of 450°C below which swelling is the governing mechanism. We will return to this issue in Section V.

Be particle interactions

We have shown that, as a consequence of internal stresses, the lifetime of Be pebbles in the LIFE neutron multiplier is likely to surpass the one-year mark called for in the LIFE design [Abbott2008]. In fact, depending on irradiation creep, Be pebbles could be kept in the reactor to Be burn-ups of up to 50% provided that the accumulated He gas can be released so as to ease swelling effects. However, many studies show that Be undergoes a drastic reduction of ductility under irradiation [Ells1964, Beeston1970, Gelles1994, Kupriyanov1995, Moons1996, Barabash2000]. While not so important in the context of a single Be particle, this property governs collisions among them, which are inevitable in configurations with free-flowing pebbles such as those envisioned in LIFE. The consensus in the literature is that Be undergoes a loss of ductility from approximately 15% to ~5% upon irradiation at 600 to 650°C. Regarding the yield strength, the variability in the data is quite large. Gelles *et al.* [Gelles1994] report values as high as 700 MPa for fluences of ~6.0×10^{21} n·cm^{-2} at 600°C. However, several measurements at 650°C have yielded lower values, between 150 and 220 MPa [Ells1964, Kupriyanov1995].

In the same fashion as Miller *et al.* [Miller1983], we estimate the contact stresses by resorting to Hertzian contact theory, which states that the maximum tensile stress at the contact point between two spheres is:

$$\sigma_{\max} = \frac{3f_c}{2\pi a^2}\left(\frac{1-2\nu}{3}\right) \quad (9)$$

where f_c is the contact force and a is the contact length:

$$a = \left(\frac{3f_c R^*}{4E^*}\right)^{\frac{1}{3}} \quad (10)$$

The effective modulus of elasticity E^* and curvature R^* are calculated for identical spheres as $E^* = \frac{E}{2(1-\nu^2)}$ and $R^*=R/2$. The criterion for failure is twofold: (i) $\sigma_{\max}<\sigma_y$, where σ_y is the fluence-dependent yield strength, and (ii) there has to be sufficient ductility to accommodate a. We consider the best and worst-case scenarios for the pebble impact conditions.

For the former, we assume that Be pebbles flow gravitationally in the (stationary) coolant and, hence, the most severe value of f_c is given by: $f_c=\rho_{\text{flibe}}Vg$, where ρ_{flibe} is the density of the coolant, $V=4\pi R^3/3$ is the pebble volume, and g is the acceleration of gravity. With $\rho_{\text{flibe}}=2.0$ g·cm^{-3} [Abbott2008] and R=15 mm, we have f_c=0.3 N, which results in values of a=0.2 mm at BOL and 0.3 mm at EOL[5]. This corresponds to $\sigma_{\max}$=0.80 MPa at BOL and 0.44 MPa at EOL. a=0.3 mm represents approximately a deformation of 2.0% in 15-mm pebbles, which is well below the ductility limit observed in irradiated Be. For its part, $\sigma_{\max}$ is insignificant compared to the yield strength measured in

[5] We have used values of E=262.7 GPa at BOL and 105.1 GPa at 30% porosity, assumed to be a reasonable EOL value for both the DU and WG-Pu cases. This results in values of E^*=131.5 and 52.6 GPa respectively.

irradiation-hardened Be. Therefore, we conclude that contact stresses due to purely gravitational pebble flow do not result in particle failure.

However, the LIFE design calls for a forced-convection coolant flowing at an average speed of 15.0 cm·s^{-1} [Abbott2008]. Thus, the worst-case scenario for pebble impact corresponds to two pebbles colliding face-on each at the velocity of the coolant. Under such circumstances, the contact force can be estimated as $f_c=2m\upsilon_0/\delta t$, where $m=\rho_{\mathrm{Be}}V$ is the pebble mass, $\upsilon_0=15.0$ cm·s^{-1} is the particle velocity, and δt is the collision time. δt can be again calculated from Hertzian mechanics as [Goldsmith1960]:

$$\ln\delta t = -\frac{1}{5}\ln v_0 + \ln\left\{\left(\frac{4.53m}{2E^*}\right)^{2/5}\left(\frac{1}{R^*}\right)^{1/5}\right\} \qquad (11)$$

which gives for our case $\delta t \approx 0.7$ and 1.0 ms for, respectively, BOL and EOL, when $\rho_{\mathrm{Be}}=1.85$ g·cm^{-3}. Hence, the contact forces at BOL and EOL are 11.2 and 7.8 N. These now result in values of $a\approx1.0$ and 1.2 mm, which represent strains of 6.7 and 8.0%. This could surpass the ductility limit of Be at high fluences and, to avoid failure, it is recommended that Be particles be coated with a stronger material that retains ductility under irradiation. This will be discussed in depth in the next section.

V. Discussion

The results show that the lifetime of Be pebbles for the DU and Pu cases, respectively, are 1.39 and 1.43 years based on a volumetric swelling limit of 30%. A critical result of this study with regard to pebble design is the establishment of an upper bound on the Be pebble radius of 15 mm, in order to prevent premature failure due to stress in excess of the tensile limit. At T>450°C, thermal creep is demonstrated to provide a full relaxation of the pebble stresses, thereby prolonging the pebble lifetimes indefinitely. Another principal outcome of our study is that the calculated mechanical performance and **Be lifetimes are practically independent of the LIFE engine design**, for the two cases considered here.

The upper bound to the pebble size is ultimately determined by also considering other engineering constraints. For example, in blanket designs where coolant flows through a "forest" of perpendicularly oriented tubes, as may be the case in LIFE, Be pebbles may be designed so as to achieve optimum flow. A case in point are the experiments performed at LLNL in 1982, where it was established that, for fission-suppressed hybrid reactors with Be pebble beds, the pebble diameter should not surpass ≈9.5 mm [Neef1982]. Of course, for the LIFE engine, the final pebble size can only be established once all the engineering has been completed, but we always recommend R<15 mm.

The long-term evolution of Be pebbles in the multiplier is dictated by the interplay between swelling and creep. Therefore, acting on the variables that control these two processes may allow engineers and designers to improve Be-particle lifetime in extreme conditions of irradiation and temperature. For example, some authors point to the fabrication procedure and material composition as a means to mitigate swelling. Sernyaev [Sernyaev1991] recommends extruded Be with O concentrations of no less than 3% and grain sizes of 10~20 microns to minimize swelling at high temperatures. Acting on the composition must be done keeping in mind the neutron multiplication capabilities of the Be, to preserve neutron balance. Alternatively, *a posteriori* treatments could be implemented based, *e.g.*, on the observations of Leenaers *et al.* [Leenaers2008], who have noted that the He accumulated in the pebbles during operation can be released by high-temperature annealings. An even better option could be to use porous Be (≥20% porosity to ensure percolation) to help release He gas during operation [Davydov2000].

A very promising material being developed mainly in Japan is metallic beryllide. Recent work shows that $Be_{12}Ti$/V/Mo intermetallics possess reduced swelling and only slightly degraded neutron multiplication properties at high fluences [Yamada2003, Kawamura2003, Mishima2007]. In particular, $Be_{12}Ti$ has been shown to undergo significantly reduced mechanical property degradation with irradiation, presumably due to the formation of a high-density of fine precipitates that act as defect sinks. In addition, it is easy to fabricate and all studies suggest that their properties are equal or superior to pure Be metal [Mishima2007]. In the context of the LIFE engine, one problem associated with these beryllides is that they are denser than the flibe (2.3 g·cm^{-3} for $Be_{12}Ti$ [Yamada2003]), which eliminates the possibility of flotation in the coolant. Only if this constraint was removed could metallic beryllides be considered a viable alternative.

With regard to creep, it is somewhat puzzling –as has been noted by Manly *et al.* [Manly1995]– that no *irradiation creep* study exists on a material as important and thoroughly studied for nuclear applications as Be. An attempt was made in 1990 at LLNL using Be pressurized tube specimens to be irradiated in FFTF/MOTA [Neef1990], although the program was cancelled before the samples were carried to the reactor. An experimental campaign is currently underway to test several Be grades at temperatures and fluences comparable to LIFE (although with 'softer' fluxes) [Hegeman2005]. This could shed light on many of the unknown issues surrounding Be at very high fluences. Based on the available data published in the literature, we are only able to provide a most conservative lifetime of 1.39 or 1.43 years, likely to be safely extended to up to two or three years. However, only after the impact of irradiation creep is reasonably established will we be able to provide a more precise lifetime window. We feel that this gap should we bridged and, to that end, **we call for an experimental effort on irradiation creep in S-65 and S-200 Be**. Another principal outcome of our study is that the calculated **Be lifetimes are practically independent of the LIFE engine design**, at least for the two cases considered here.

The above considerations are based solely on isolated pebbles developing internal stresses. However, we have seen in Section IV that inter-particle collisions could be a source of failure at high fluences, after Be ductility has been degraded to less than 5%. In such circumstances, it is necessary to coat each pebble with a material that maintains sufficient ductility during the lifetime of the particles. This is easily achieved by many transition metals, although, ideally, they should be chemically compatible with Be and have low activation. Among these are Ta, W, and V [Lindau2005], although, interestingly, not stainless steel [Beeston1970]. Since the main purpose of the coating is not to provide mechanical integrity but to avoid direct Be-Be contact, very thin shells could be used. However, fabricating these shells may present a significant technological challenge and may lead to defective (high crack concentration) coatings. Nevertheless, a distribution of cracks could aid in degassing the Be particles for extended lifetime purposes.

Furthermore, the pebble coating must resist corrosion by the flibe coolant, which, among the candidate materials mentioned, **W is capable**. The use of a W coating could also serve as a containment barrier once the metallic Be crumbles at swellings of 30% or higher. This limit could be extended by releasing the trapped He gas through an unspecified number of small holes punched through the protective coating. This may expose the Be to corrosion by the flibe, which has been shown to be a important [Hara2006]. However, the use of redox agents [Petti2006] in conjunction with the porosity of the Be pebble should help in mitigating this issue. For flotation, the thickness of the W layer must be such that the density of the combined Be/W pebble not surpass that of the flibe. For W, this condition is guaranteed for thicknesses of no more than 40 μm (see Appendix for this calculation), which, again, may be technologically unrealistic with current fabrication methods. Thus, in addition to the requirements laid out above, we recommend that 20%-porous Be be used to allow thicker W coatings. From eq. (A2), this would then result in a thickness of 145 μm.

As we have pointed out, if pebble flotation in the flibe is not a constraint, one could use thicker W coatings, or $Be_{12}Ti$ as the multiplier material. Studies show that $Be_{12}Ti$ possesses a lower ductility than pure Be [Kawamura2003], although there are also indications that the beryllide retains ductility with irradiation much better. There is some evidence as well that beryllides are more impervious to corrosion than pure metallic Be [Mishima2007]. However, until these issues are more conclusively studied, we propose that $Be_{12}Ti$ particles be coated with chemically compatible ferritic/martensitic (F/M) or oxide-dispersed strengthened (ODS) steels, to take advantage of their excellent irradiation resistance. Subject to fabrication limitations, the minimum thickness in the case of a steel coating is then given by the corrosion rate in flibe of 5 μm per year below 700°C according to Zinkle and Ghoniem [Zinkle2000]. However, a double coating of steel/W could be a conceivable solution as well if steel corrosion is demonstrated to become a major issue.

Whether the design chosen is Be/W or $Be_{12}Ti$/steel pebble, the life-limiting factor is no longer dictated by eqs. (1), but by the radiation resistance of the coating. For example, for the $Be_{12}Ti$/steel case, our calculations show that the damage rate in Fe in the Be multiplier is 18 dpa per year for both LIFE designs. Assuming the state-of-the-art damage resistance for ODS steels of ≈110 dpa at 600°C [Toloczko2004], the coated pebbles could last more than six years before failure. Combining the conditions of corrosive thinning with irradiation resistance, we arrive at a minimum ODS coating thickness of at least six times the corrosion yearly rate, although for safety we propose 10 times. For the standard rate of 5 μm/year that yields 50 μm.

For W (5 dpa per year in LIFE's Be blanket) the uncertainties are higher as there are no experimental data above 5 dpa [Matolich1974]. However at 600ºC and fast fluences in excess of 10^{22} $n\cdot cm^{-2}$ the swelling observed was always less than 0.5% [Matolich1974], which leads us to speculate that it could also safely withstand 6 years of irradiation in the LIFE Be blanket. We emphasize that, in this paper, we are not taking into account fabrication limitations directly, which we understand may be the stumbling block regarding Be pebble engineering.

VI. Conclusions

On the basis of our calculations and the available material property data, **we propose for the LIFE engine buoyant Be pebbles <15 mm in radius containing ≈20% porosity coated with no more than 145 μm of W.** For alternative engine designs where flotation in the flibe is not a requirement, **we propose $Be_{12}Ti$ pebbles coated with a shell of ODS steel with a minimum thickness of 50 μm**. This value is obtained assuming a corrosion rate of 5 microns per year, which will be updated in upcoming studies for the LIFE engine. Both designs include the presence of holes in several locations to allow the release of He gas.

The calculated lifetimes for uncoated, 15-mm-radius Be pebbles are ≈1.4 years, pending on more reliable data for irradiation creep in metallic Be. **The lifetime of the coated pebbles is estimated to be of a minimum of 6 years. The design temperature window for all pebbles proposed is 450ºC<T<700ºC**.

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Appendix: Calculation of W and ODS steel coating thicknesses for encapsulated Be pebbles.

Pebble floatability in flibe is ensured by solving the following inequality:

$$\frac{4}{3}\pi\rho_c\left(R_c^3 - R_{Be}^3\right) + \frac{4}{3}\pi\rho_{Be}R_{Be}^3 < \frac{4}{3}\pi\rho_{\text{flibe}}R_c^3 \quad \text{(A1)}$$

where R_c and R_{Be} are the radii of the total coated pebble and the Be core respectively, and ρ_c, ρ_{Be} and ρ_{flibe} are the corresponding coating, Be and flibe densities. Eq. (A1) reduces to the following condition:

$$R_c < R_{Be}\left(\frac{\rho_c - \rho_{Be}}{\rho_c - \rho_{\text{flibe}}}\right)^{1/3} \qquad \text{(A2)}$$

Solving eq. (A2) for R_{Be}=15 mm, ρ_{flibe}=2.00 g·cm^{-3}, ρ_{Be}=1.85 g·cm^{-3} (fully-dense Be), and ρ_c=19.30 g·cm^{-3} (W), we have $R_c \approx 15.04$ mm, *i.e.* coatings no larger than 40 microns. For 80%-dense Be (20% porosity), we obtain $R_c \approx 15.15$ mm.

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